Response to reviewer #1

Authors appreciate reviewer's thoughtful comments and suggestions, which are greatly helpful for us to improve our manuscript. The manuscript has been revised to accommodate the reviewer's comments.

General Comments:

The manuscript describes the implementation of the trajectory-grid technique into the popular CMAQ regional air quality model. It demonstrates the use of this upgraded model in simulating transport of CO during the KORUS-AQ campaign, demonstrating that it could be used to identify likely sources of observed pollution. The paper is fundamentally suitable for GMD, as it describes an interesting new diagnostic technique for a widely-used model. The trajectory-grid method is interesting, and the integration of a Lagrangian technique into an Eulerian model has the potential to bring valuable new insights into air quality modeling. However, I believe that the authors overstate their conclusions in ways which are not necessarily supported by their work. I also find that the paper lacks some important methodological details. I would therefore recommend that the paper undergo some revisions before being accepted for publication in GMD.

Major comments: My biggest concern is that certain aspects of the method seem to be either incompletely described or just incomplete. In particular:

Comment) If parcels are spawned in cells with the concentrations of the closest packet, how is this approach mass conservative? Chock et al (2005) did claim that the T-G method is mass conserving, but they point out that this is not the case when diffusion is simulated. Similarly, it is not clear to me how the spawning and pruning processes (lines 105-109) could occur without either incurring mass conservation errors or acting to artificially diffuse the concentration distribution.

Response) Thank you for your insightful comment. As we mentioned in the manuscript, Line 96-99: "Since we can use the TG method to calculate the concentration from an ordinary differential equation, it is mass conserving, monotonic, and accurate. In the diffusion step, however, interpolation errors occur, but they are typically considerably smaller than Eulerian advection errors (Chock et al., 2005)." Therefore, your comment is valid, and we addressed the error caused by pruning and spawning by adding another sentence in Line 125-126 "The limitation of this packet management approach, however, is that it incurs mass conservation by adding minor interpolation errors."

Comment) The approach described is designed with long range transport in mind, but I could not find any description of how convection is treated. Convective transport could significantly change the path taken by an advected air parcel, and is not trivial to simulate in a Lagrangian framework. How is this phenomenon dealt with in this model? I could not see any reference to it in Figure 2.

Response) Based on CMAQ's Science document, Chapter 11: "The resolved clouds have been simulated by the MM5 to cover the entire grid cell. No additional cloud dynamics are considered in CMAQ for this cloud type since any convection and/or mixing would have been resolved and considered in the vertical wind fields provided by MM5." Also, we checked the WRF Kain-Fritsch Model: "…, momentum transport in convective clouds is crudely simulated by assuming conservation of momentum in convective clouds…"

$$\left.\frac{\Delta\bar{u}}{\Delta t}\right|_{conv} = \frac{1}{\Delta p}\left[(\omega_{u2}+\omega_{d2})\bar{u}_2 - (\omega_{u1}+\omega_{d1})\bar{u}_1 + (\epsilon_u+\epsilon_d)\bar{u}_m - \delta_u u_{um} - \delta_d u_{dm}\right]$$

$$\left.\frac{\Delta\bar{v}}{\Delta t}\right|_{conv} = \frac{1}{\Delta p}\left[(\omega_{u2}+\omega_{d2})\bar{v}_2 - (\omega_{u1}+\omega_{d1})\bar{v}_1 + (\epsilon_u+\epsilon_d)\bar{v}_m - \delta_u v_{um} - \delta_d v_{dm}\right]$$

TG advection in C-TRAIL solves the transport equation in a three-dimensional format; therefore, the convective transport is already considered in vertical wind fields. Sub-grid clouds convection, however, will be considered in a future study.

Comment) I disagree with one of the premises of this study. On lines 46-48, it is claimed that backtrajectory modeling for source-receptor estimation is "not widely accepted because it is unable to determine whether an originated air mass is polluted or non-polluted". Given the widespread use of back-trajectory modeling for this exact purpose, this statement does not seem justified. Furthermore, it seems to neglect that the same issue exists with the C-TRAIL approach, but in reverse. Unless an simulated air mass exactly coincides with the monitor location at the exact time of an observation, one cannot be certain that it contributed to that specific reading.

Response) Thank you for your comment. You are right about the widespread use of back-trajectory modeling for finding the originated air mass. In this study, however, our approach is not based on observations. For the domain of our study, we are using an emission inventory platform for a gridded emission inventory. With the help of this gridded domain and use of the TG method in following air parcels, we are attempting to link the receptor air parcel with the originated grid. Therefore, we determine how much pollution the originated grid provided by its emission and concentrations. The C-TRAIL approach is a forward trajectory model, with millions of air parcels moving in the domain, some of which will pass through (or land in) the receptor location. Unlike other conventional trajectory methods, this approach uses gridded emissions/concentrations across the domain (which is not the case for observation sites; that is, they are available only where the observation site is located). Therefore, we think that the C-TRAIL approach has some benefits. The limitation of this approach, however, is the uncertainty of the emission inventory and/or model concentrations (which is why we provided the correlation and IOA values).

Comment) Line 198 – what does it mean to "reach Seoul" (altitude)? What is the minimum altitude that a packet must reach to be considered as "reaching" Seoul? This seems important – if the altitude is too great, then air packets which are simply passing over Seoul (and therefore irrelevant to its air quality) will be incorrectly labeled as contributions. Furthermore it would be strange to compare two parcels which arrived at very different heights.

Response) Thank you for your comment. The word "reaching," as you mentioned, does not always mean affecting air quality issues, but as we have shown in the supplemental Figure S8 referring to heights, the air parcels over Seoul are at the 1.5km and lower more than 80% of the time. Furthermore, in our explanations, we divided the air parcels that come from higher altitudes from those which are at a lower altitude (below the boundary layer height ~ 1km) to determine the impact on LRT. You can also find this information in Figures 7-10 (the color-bar), in which most of the air parcels are blue. We agree with your point, so we made some modifications to the manuscript.

Line 199-200: "… different packets from different altitudes (from below 1 km to almost 10 km)… "

Line 226: "**Error! Reference source not found.**(a) presents the general path of all packet trajectories reaching the Seoul area at different altitudes at 9:00 AM LT throughout May 2016."

Line 215-216: "The concentrations of near-surface packets tend to fluctuate more than those of high-altitude packets (Figure S7)."

We also added a plot in a supplementary document for height change of 4 packets reaching Seoul to show their altitudes:

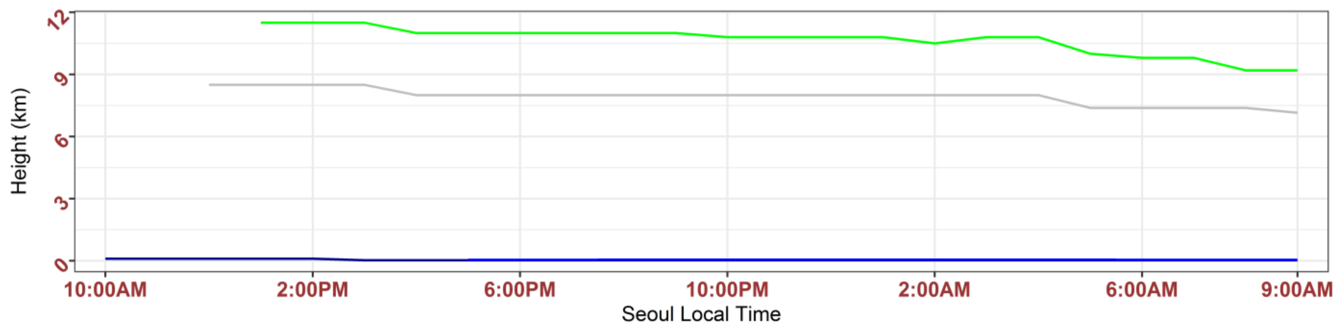

Figure S7: Changes in the height (km) of four aged packets moving toward Seoul from source points

Comment) Throughout the paper, it is claimed that the model is "accurate" (line 296), "ideal" (line 302), and "validated" (line 11). However, the main validation is shown in Figure 4, and – unless I have misunderstood – this just shows that the base CMAQ model (since it does not seem that the TG modifications change the behavior of the base model) can reproduce some of the observed behavior. With that in mind: 1) Why does the model seem to consistently underpredict the observed values? I note that the authors do state as much for the DWP and SP, but they also state "very high correlation" during the EPP. While technically true that the index of correlation is higher during EPP than DWP or SP, this seems misleading when the root mean square error is still 68.7 ppbv. 2) Why does the model appear to have a hard minimum concentration of 50 ppbv CO (see figure 4a)? 3) In what sense is the TG approach "validated" here? If no specific validation of the C-TRAIL approach is given (e.g. by comparing to continuous satellite observations), I would recommend that the authors instead simply state that this is a new diagnostic technique for an existing model. I applaud the authors for bringing up some of the shortcomings of the method in their conclusions (lines 305-306). However, I recommend that they put a more comprehensive discussion of these limitations in the main text.

Response) 1) The underprediction of the standard CMAQ model vs. observations is mostly due to the quality of the emission inventory over East Asia. Several studies have obtained similar results from CMAQ over East Asia. When providing the correlation, however, we also calculate the IOA (index of agreement), which combines the correlation and mean bias errors. The IOA for EPP shows a 0.88 value, which is generally high for the IOA. Your concern regarding the underprediction will be addressed by data assimilation and inverse modeling techniques in future studies.

2) The model here is compared to the KORUS-AQ aircraft observation datasets provided in the KORUS-AQ website. The dataset covers several species at different altitudes during the KORUS-AQ period, May and June 2016. The minimum of about 50ppbv comes from observations that established this minimum from measurements. We also compared our model's CO predictions with surface measurements for stations across South Korea (Table S3, Figure S5). The results showed the discussed underprediction, which we think is because of uncertain data acquisition for ground measurements (error ±100 ppbv for CO), uncertain emission inventory over East Asia, and model's uncertainty in chemistry modeling.

3) We agree with the reviewer that this model is a new diagnostic technique for the CMAQ model, and we added a related comment in the Discussion section. In the following version of C-TRAIL in a future study, we will implement all CMAQ modules in C-TRAIL and compare these two models with observations.

Line 11-12: "The diagnostic output from C-TRAIL accurately identifies the origins of pollutants."

Line 190-191: “Because C-TRAIL is a diagnostic tool derived from CMAQ, both a Lagrangian output and CMAQ standard Eulerian output are available after each run.”

Comment) Minor comments Line 29 – the words in HYSPLIT should be capitalized (i.e. “Hybrid Single-Particle Lagrangian Integrated Trajectory”)

Response) Thanks for your comment. Revised.

Comment) The manuscript contains numerous grammatical errors. This does not affect the quality of the science or whether it should be accepted, but I would recommend that the authors take another pass through to try and improve this.

Response) We rewrote some sections due to some grammatical errors. Thank you for your suggestion.

Comment) I noticed that the code is made available only by request. While this is fine, I would encourage the authors to consider making the code more freely available (e.g. on GitHub) unless they are unable to do so because of some specific restriction.

Response) Based on reviewer’s suggestion we provided this code in Github. You can find the link at the Code Availability section.

Line 348-349: “**Code Availability.** The C-TRAIL version 1.0 is available for non-commercial research purposes at https://github.com/armanpouyaei/C-TRAIL-v1.0.”

Comment) I recommend that the abstract be toned down.

Response) Thanks, Revised.

Comment) Several claims do not seem to be meaningful (e.g. on line 6, how is the Lagrangian output “reliable” or “comprehensive”?) and others are confusing (e.g. I’m not sure what is meant by “real polluted air masses” – line 8).

Response) We addressed these types of claims in the manuscript. Thanks.

Comment) More generally, I note that the term “real polluted air mass” turns up many times. I suggest this is changed to simply “polluted air mass” given that these are still all simulation results. Even if the pollution was observed at some point, the model is performing a simulation which will include errors.

Response) Addressed.

Line 8: “…for validating the source-receptor direct link by following polluted air masses.”

Line 63: “: (1) to provide a direct link between polluted air masses from sources and a receptor”

Comment) On lines 309 and 310, the authors call the model “efficient”. However, I did not see any quantification of this in the paper. I suggest that this claim is removed. Alternatively the authors could consider giving a specific estimate of the computational overhead associated with C-TRAIL.

R) Thanks for the comment. Based on your suggestion, we removed this claim.